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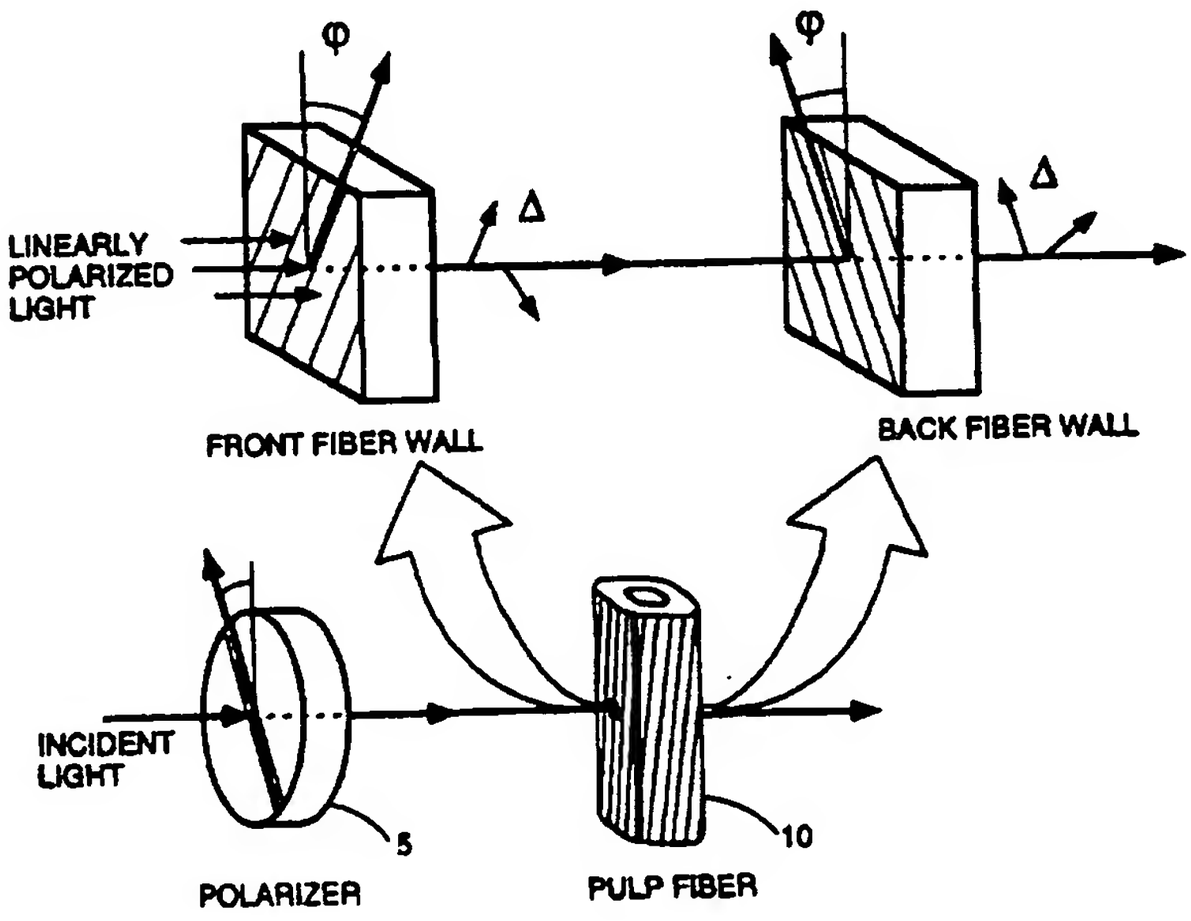

(57) Abstract

The presented invention provides a new method and device for determining the fibril angle and the relative phase retardation of single, intact pulp fibers. The new method is based on the intensity quotient ellipsometry and uses the multi-wavelength principle to determine the measurement results. The method of the invention permits a two-fiber-wall measurement, therefore requires no sample pretreatment. Compared with the existing ones, the new method is simple, fast, more accurate and non-destructive of the fiber material. A device for determining the fibril angle and the relative phase retardation of single, intact pulp fibers in accordance with the present invention comprises a light source, a polarization-optical image system, a detector and an image-processing unit.

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METHOD AND DEVICE FOR DETERMINING THE ORIENTATION ANGLE OF THE OPTICAL AXIS AND THE RELATIVE PHASE RETARDATION OF A BIREFRINGENT SPECIMEN

FIELD OF THE INVENTION

This invention relates to a method and device for determining the orientation angle of the optical axis and the relative phase retardation of a birefringent specimen, preferably the fibril angle and the relative phase retardation of intact pulp fibers.

BACKGROUND OF THE INVENTION

A wood fiber consists of a primary wall enveloped in lignin to form the middle lamella, and three secondary walls, S_1, S_2 and S_3 layers (e.g. ref. R. D. Preston, The physical biology of plant cell walls, (Chapman and Hall Ltd., 1974)). All the three secondary walls are concentric and composed of cellulosic micro-fibrils, embedded in an amorphous matrix of hemicelluloses and lignin. The most important wall is the middle secondary wall S_2 because it contains most of the fiber material (80-95%) (see D. H. Page, "A method for determining the fibrillar angle in wood tracheids," Journal of Microscopy 90, 137-143 (1969) and R. E. Prud'homme and J. Noah, "Determination of fibril angle distribution in wood fibers: a comparison between the x-ray diffraction and the polarized microscope methods," Wood and fiber 6, 282-289 (1975)) so that a wood pulp fiber can be approximately described by the S_2 layer. The crystalline microfibrils of the S_2 layer trace a steep spiral around the fiber axis so that the microfibrils of the front and back walls of the S_2 layer are crossed. The angle between the fibrillar direction and the fiber axis is termed the fibril angle or micellar angle of the fiber.

The fibril angle is closely related to the mechanical properties of the fibers, such as the strength, the elastic modulus and the shrinkage (see R. E. Mark and P. P. Gillis, "The relationship between fiber modulus and S_2 angle," Tappi 56, 164-167 (1973); F. El-Hosseiny and D. H. Page, "The mechanical properties of single wood pulp fibres: theories of strength,"

Fibre Science and Technology 8, 21-30 (1975); D. H. Page, F. El-Hosseiny, K. Winkler, and A. P. S. Lancaster, "Elastic modulus of single wood pulp fibers," Tappi 60, 114-117 (1977); and D. H. Page and F. El-Hosseiny, "The mechanical properties of single wood pulp fibres. Part VI. Fibril angle and the shape of the stress-strain curve," Journal Pulp and Paper Science, TR 99-100 (1983). To determine the fibril angle, many methods have been developed and used, for example striation observation, angle of the slit pits, iodine staining, X-ray diffraction and polarized-light microscopy. The first three techniques are tedious and only applicable to some wood species (see, e.g. Preston). In general, the X-ray diffraction method is suitable for giving a measure of the mean fibril angle of a piece of wood consisting of a few hundred fibers. Unfortunately, however, the interpretation of the diffraction patterns to obtain the fibril angle has not been clearly established (B. A. Meylan, "Measurement of microfibril angle by x-ray diffraction," Forest Prod. J. 17, 51-58 (1967) and T. Paakkari and R. Serimaa, "A study of the structure of wood cells by x-ray diffraction," Wood Sci. Technol. 18, 79-85 (1984)). Additionally, the application of the X-ray technique relies heavily on fiber geometry that is uncertain (see e.g. R. E. Prud'homme and J. Noah).

Polarized-light microscopy has been employed for many years to measure the fibril angle. The main difficulty with this technique is that a single, whole fiber (represented by the S_2 layer) has two walls. This made it very difficult to measure the whole fiber without pretreatment. In fact all the existing polarizing microscope methods, as described by Preston, Page, Prud'homme et al, C. M. Crosby and R. E. Marke, "Precise S_2 angle determination in pulp fibers," Svensk Papperstidning 17, 636-642 (1974); C. M. Crosby, C, De Zeeuw, and R. Marton, "Fibrillar angle variation in red pine determined by Sénarmont compensation," Wood Sci. and Technol. 6, 185-195 (1972); Ruen C, Tang, "The Microfibrillar orientation in cell-wall layers of virginia pine tracheids," Wood Science 5, 181-186 (1973); and Lawrence Leney, "A technique for measuring fibril angle

using polarized light," Wood and Fiber 13, 13-16 (1981), are applicable only to one fiber-wall measurement. The most commonly used m.e.p. (major extinction position) method described by Preston needs to cut one wall away and leave the other one available for examination. This is not a simple procedure because the fibers are usually only several tens of µm wide. The variant m.e.p. method described by Page allows one fiber wall not to be removed from the other. In this method, mercury droplets are injected into the lumen under high pressure. The deposited mercury droplets will function as small mirrors, reflecting the polarized light so that it passes through only one wall. Because of the one-wall measurement, the existing polarized-light methods destroy the fiber material and require tedious and time-consuming pretreatment of samples. Any physical or chemical treatment will after the fiber geometry or properties to some extent. Besides, the existing methods are inaccurate.

A plane polarized light when entering the S_2 layer is split into two orthogonal components travelling at different velocities and they have a relative phase retardation when emerging from the S_2 layer. The theory predicts that the phase retardation is proportional to the fiber-wall thickness, another basic wood fiber quantity. The wall thickness is related to the fiber flexibility and directly tied to the coarseness value of the fiber. The wall thickness is usually measured by cutting fibers into very thin sections. A non-destructive measurement of the wall thickness is possible if the phase retardation is known. However, so far the retardation can be measured only for fibers one of whose two walls is removed and for which the fibril angle has been determined (ref. e.g. Preston).

SUMMARY OF THE INVENTION

It is therefore an object of this invention to provide a new method and device for measuring the fibril angle and the phase

retardation of a birefringent specimen, preferably of single pulp fibers without any sample pretreatment.

Another object of the invention is to provide a method that should permits a nondestructive, simpler and more accurate measurement of the fibril angle and the phase retardation of a birefringent specimen, preferably of pulp fibers.

A further object of the invention is that a device for measurement of pulp fibers according to the invention should be as simple as possible so that it can be easily and inexpensively manufactured.

The present invention provides a new polarized-light method as the solution for determining both the fibril angle and phase retardation of single, intact wood pulp fibers. A device used in this method comprises a light source, a polarization-optical image system in connection to a detector and an image-processing unit. The image system consists of a polarizer and an analyzer and the fiber to be measured is located between them. For this method the polarizer is fixed with an angle, most preferably an angle of 45°, relative to the axis of the fiber or fiber segment to be measured and the emergent light from the fiber or fiber segment is measured by rotating the analyzer.

The measurement method of the invention is the first one that permits two fiber-wall measurement of pulp fibers. Due to its two-wall measurement technique the method requires no sample preparation. In contrast to existing ones, besides, the method of the invention allows a quantitative determination of the fibril angle and the phase retardation. Therefore it is relatively simple, fast, basically more accurate than the existing ones and non-destructive of the fiber material. The method will make it much easier to investigate the fiber properties reflected in changes in the fibril angle or/and the retardation.

The measurement method of the invention, which has been experimentally demonstrated, provides two approaches for determination of the fibril angle and relative retardation where either the light intensity is detected at four special analyzer positions for a simpler and quicker measurement or the light intensity within one period (180°) is registered for a higher accuracy.

For the method of the invention the same measurement proceedings have to be repeated for at least two wavelengths to be able to determine the measurement results. The wavelength change can be easily realized by employing a manochromator or a normal polychromatic light source equipped with a set of at least two wavelength filters. Moreover, to reduce the influence of possible scattering effect, the measurement point is preferably chosen in the middle region of the fiber.

In some retardation ranges, when applying the method of the invention, the error can be very large. This limitation does not severely restrict the applicability of the method, because it is always possible to avoid the retardation range, in which the errors are too large, by changing the wavelength. To recognize a critical case it is only necessary to check the amplitude of the intensity variation generated by rotating the analyzer. If the amplitude is too large new wavelength should be chosen to make the intensity vary more smoothly.

Besides the wood pulp fiber, in principle, the measurement method of the invention is suitable for other cellulose fibers such as cotton, ramie and flax fibers.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 illustrates the model for describing the polarization transmission of a whole pulp fiber used in the method of the invention, the two fiber walls are represented by two optical linear retarders of the same relative retardation Δ and the same orientation angle φ (fibril angle) with opposite senses.

FIG. 2 is a schematic diagram of a device for determining the fibril angle and the relative phase retardation of single, intact pulp fibers in accordance with the present invention.

FIGS. 3a and 3b are the polarizing micrograph (λ=600 nm) of unbleached commercial pine kraft pulp fiber and the fiber segment (420x) measured by using the method of the invention, the two marked points in the centre region were measured.

FIGS. 4a, 4b, 4c and 4d illustrate the results for the fibril angle φ and the retardation Δ of the pulp fiber in FIG. 3b at point 1 and point 2 versus wavelength λ.

FIG. 5 shows the detected intensity dependence I(A) (λ=600 nm) at point 1 of the fiber shown in FIG. 3b.

FIGS. 6a and 6b are the polarizing micrograph (λ=600 nm) of an unbleached laboratory-made pine kraft pulp fiber and the fiber segment (420x) measured by the method of the invention, the two marked points in the centre region were measured.

FIGS. 7a, 7b, 7c and 7d illustrate the results for the fibril angle φ and the retardation Δ of the pulp fiber in FIG. 6b at point 1 and point 2 versus wavelength λ.

FIGS. 8a and 8b are the polarizing micrograph (λ=600 nm) of a bleached commercial pine kraft pulp fiber and the fiber segment (420x) measured by the method of the invention, at the two marked points in the centre region were measured.

FIGS. 9a, 9b, 9c and 9d illustrate the results for the fibril angle φ and the retardation Δ of the pulp fiber in FIG. 8b at point 1 and point 2 versus wavelength λ.

DESCRIPTION OF THE PREFERRED EMBODIMENT

To develop a quantitative method that permits a nondestructive measurement of the fibril angle and the phase retardation of

pulp fibers using the polarized-light technology, first it is necessary to describe the polarization transmission property of a whole fiber. As pointed out, a whole fiber has two fiber walls whose microfibril directions are crossed. In general, it can be further assumed that the fiber-wall thickness in the region under study is constant, the microfibrils lie parallel to each other and the refractive index field formed by the microfibrils is homogeneous. Moreover, it is always possible to examine the middle region of a fiber where the influence of light scattering from the fiber wall should be small to be ignored. Under these conditions, the present invention provides a model for describing a whole pulp fiber (FIG. 1). In FIG. 1 a linearly polarized light generated by a polarizer 5 is incident on a pulp fiber 10 and the two opposite fiber walls of the fiber 10 are represented by two optical linear retarders with crossed axes in cascade. These two retarders have the same relative retardation Δ, which is proportional to the thickness of the fiber wall and the birefringence of the wall material. The optical orientation angles of these two retarders have the same value as the fibril angle φ of the fiber, but with opposite senses. By using the Jones-matrix formalism, developed by R. C. Jones, "A new calculus for the treatment of optical systems," Part I-III. J. Opt. Soc. Amer. 31, 488-503 (1941) and further described by R. M. A. Azzam and N. M. Bashara, Ellipsometry and Polarized Light (North-Holland, New York, 1979); P. S. Theocaris and E. E. Gdoutos, Matrix Theory of Photoelasticity (Springer-Verlag Berlin, Heidelberg, New York, 1979); and Amnon Yariv and Pochi Yeh, Optical Waves in Crystals: Propagation and Control of Laser Radiation (John Wiley & Sons, 1984), therefore, the transmission property of a fiber with the fiber axis as the reference axis can be described by its Jones-matrix

$$\mathbf{T}_S = \mathbf{R}(\varphi)\begin{bmatrix} e^{-j\Delta/2} & 0 \\ 0 & e^{j\Delta/2} \end{bmatrix}\mathbf{R}(-\varphi)\,\mathbf{R}(-\varphi)\begin{bmatrix} e^{-j\Delta/2} & 0 \\ 0 & e^{j\Delta/2} \end{bmatrix}\mathbf{R}(\varphi), \qquad (1)$$

with the rotation matrix

$$R(\varphi) = \begin{bmatrix} \cos\varphi & \sin\varphi \\ -\sin\varphi & \cos\varphi \end{bmatrix}. \quad (2)$$

Furthermore, T_s can be written as

$$T_S = \begin{bmatrix} a & b \\ c & d \end{bmatrix}, \quad (3)$$

where

$$a = \sin^2(2\varphi) + \cos(\Delta)\cos^2(2\varphi) - j\cos(2\varphi)\sin(\Delta), \quad (4a)$$

$$b = -\sin(4\varphi)\sin^2\left(\frac{\Delta}{2}\right), \quad (4b)$$

$$c = -b = \sin(4\varphi)\sin^2\left(\frac{\Delta}{2}\right) \quad (4c)$$

$$d = \sin^2(2\varphi) + \cos(\Delta)\cos^2(2\varphi) + j\cos(2\varphi)\sin(\Delta). \quad (4d)$$

Based on equations (3) and (4) and using the principle of the intensity quotient method of the transmission ellipsometry described by W. Holzapfel and C. Ye, "Transmission ellipsometry of Δ and φ based on intensity quotient measurements," Optik 91, 53-60 (1992) and C. Ye, "Untersuchungen zur photoelastischen Polarisationsmodulation optischer Signale," doctoral thesis (Kassel University, Kassel, Germany, Verlag Shaker Aachen, 1992, ISBN 3-86111-201-9), the method of this invention is developed, whose principle will be described below in more detail with reference to the device of the invention.

FIG. 2 schematically illustrates a device in accordance with this invention for determining the fibril angle and the relative phase retardation of single, intact pulp fibers. The device comprises a light source unit 1, a polarization-optical image system 4, a detector 11 and an image-processing unit 12. The light source unit 1 can be an optical monochromator or a tunable laser, which can provide monochromatic or quasi-mono-

chromatic, preferably visible, radiation of at least two wavelengths. In this embodiment the light source unit 1 comprises a normal light source 2 which generates a light beam having a broad spectrum and a filter convector 3. The filter convector is preferably automated to sequentially insert N wavelength filters 3_1, 3_2, ... 3_N ($N \geq 2$) into the path of the light beam generated by the light source 2 so that the light source unit 1 sequentially feeds a monochromatic or quasi-monochromatic radiation of wavelength λ_1, λ_2, ...λ_N ($N \geq 2$) to the polarization-optical image system 4.

The polarization-optical image system 4 comprises a polarizer 5, a microscope condenser 6, a workstage 7, a microscope objective 8 and a rotatable analyzer 9. A pulp fiber 10 to be measured is installed on the workstage 7. As shown in FIG. 2, the light beam from the light source unit 1 with constant intensity 10 enters the polarization-optical image system 4 and it is first linearly polarized by the polarizer 2 (azimuth P). The linearly polarized light is focused to the fiber 10 on the workstage 7 through condenser 5. By rotating the workstage 7 the axis of the fiber 10 can be oriented with a certain angle relative to the transmission axis of the polarizer 5. The workstage 7 can be replaced by a sampling system comprising a capillary (e.g. Kajaani FS-200), which is located in a plane perpendicular to the light direction and oriented with a desirable angle relative to the polarizer 5. The fibers to be measured will be one by one passed through the capillary at speed slow enough so that the effect due to the movement of the fibers can be ignored. The fiber 10 to be measured is magnified and imaged by the objective 8, after passing through the analyzer 9, to the detector 11. The detector 11 can be either a CCD camera or a microscope video camera. The light emergent from fiber 10 is, in general, elliptically polarized and analysed by rotating the analyzer 9 (azimuth A). The light intensity at different analyzer positions are recorded by the detector 11. The obtained light intensity data by the detector 11 are digitized by image processing unit 12, which is interfaced to a computer where the results for the fibril

angle and the relative phase retardation are calculated by using the method described below.

With the help of equations (3) and (4) the electric field vector E of the light beam behind the analyzer can be given. It follows

$$\mathbf{E} = \sqrt{I_0}\begin{bmatrix}\cos A & \sin A\\ 0 & 0\end{bmatrix}\begin{bmatrix}a & b\\ c & d\end{bmatrix}\begin{bmatrix}\cos P\\ \sin P\end{bmatrix}. \qquad (5)$$

After some calculations the transmitted light intensity $I(A)=\mathbf{E}\cdot\mathbf{E}^*$ ($\mathbf{E}^*$: the complex conjugate of E) can be obtained as follows:

$$\begin{aligned} I(A)=I_0\Big\{&\Big[c^2\cos^2 P+ad\sin^2 P+\frac{c}{2}(a+d)\sin 2P\Big]\sin^2 A\\ &+\Big[ad\cos^2 P+b^2\sin^2 P+\frac{b}{2}(a+d)\sin 2P\Big]\cos^2 A\\ &+\frac{1}{2}\Big[c(a+d)\cos^2 P+b(a+d)\sin^2 P+\frac{1}{2}\left(a^2+d^2+2bc\right)\sin 2P\Big]\sin 2A\Big\}. \end{aligned} \qquad (6)$$

According to the method of the invention, the polarizer 5 is fixed with a certain angle, most preferably of 45°, relative to the axis of the fiber 10 and the light intensity behind the analyzer is detected by rotating the analyzer 9. Then, by calculating the quotients based on the measured intensity data, useful information about the fibril angle and the retardation of the fiber 10 will be obtained. Because in many cases the fibers are not straight, the concept of the local fiber axis has to be applied in practice. For a curved fiber the axis of a fiber segment which is straight enough is a local axis of this fiber. By using equations (4a)-(4d) and for P=45° equation (6) can be simplified to

$$I(A) = \frac{I_0}{2}\{1 + T_1 \sin 2A + T_2 \cos 2A\}, \quad (7)$$

where

$$T_1 = 1 + 8\cos^2(2\varphi)\,\sin^2\left(\frac{\Delta}{2}\right)\left[\cos^2(2\varphi)\,\sin^2\left(\frac{\Delta}{2}\right) - 1\right], \quad (8a)$$

$$T_2 = 2\sin(4\varphi)\,\sin^2\left(\frac{\Delta}{2}\right)\left[2\cos^2(2\varphi)\,\sin^2\left(\frac{\Delta}{2}\right) - 1\right]. \quad (8b)$$

For the method of the invention, there are two alternative ways available for determining φ and Δ. Because the harmonic part of I(A) depends only on φ and Δ, the least-square fitting method can be used to obtain the best estimates for these parameters. To do this, the intensity dependence I(A) within one whole period needs to be registered. From the intensity data obtained a harmonic curve fitting the intensity data is generated by varying the values for φ and Δ in equation (7). After determining the fitting curve, the best estimates for both φ and Δ are obtained.

Another approach for determining φ and Δ is based on determining T_1 and T_2 in equations. (8a) and (8b) by measurement. Actually T_1 and T_2 are the intermediate quantities connecting the polarization parameters to be measured, i.e. φ and Δ, and the intensity values required for measuring them. From equation (7) the following relations can be written:

$$T_1 = \frac{I(A = 45°) - I(A = 135°)}{I(A = 45°) + I(A = 135°)} \quad (9a)$$

and

$$T_2 = \frac{I(A = 0°) - I(A = 90°)}{I(A = 0°) + I(A = 90°)}. \quad (9b)$$

For this method intensity values at only four special positions of analyzer 5 are required. Note that in equations (9a) and (9b) the principle of the quotient calculation has been applied. It should be pointed out that through the quotient calculation the influence of intensity fluctuation and other errors are compensated. Equations (9a) and (9b) indicate that T_1 and T_2 can be determined by detecting the intensity at the analyzer positions A=0°, 45°, 90° and 135° relative to the fiber axis. In fact, T_1 and T_2 can also be obtained by measuring the light intensity at four analyzer positions A_1, $A_2=A_1+45°$, $A_3=A_1+90°$ and $A_4=A_1+135°$. After ascertaining T_1 and T_2, equations (8a) and (8b) can be considered as a pair of equations with only φ and Δ as unknown variables. In fact, equation (8a) is quadratic with respect to the expression $\cos^2 2\varphi \cdot \sin^2(\Delta/2)$ and has two solutions. Substituting the two solutions for the expression $\cos^2 2\varphi \cdot \sin^2(\Delta/2)$ into equation (8b), respectively, the representations for φ and Δ can be obtained as follows:

$$\begin{cases} \operatorname{tg} 2\varphi_1 = \dfrac{-2T_2}{\sqrt{2(1+T_1)}\left(2-\sqrt{2(1+T_1)}\right)} \\ \operatorname{tg} 2\varphi_2 = \dfrac{2T_2}{\sqrt{2(1+T_1)}\left(2+\sqrt{2(1+T_1)}\right)} \end{cases} \qquad T_1 \neq \pm 1 \qquad \text{(10a and b)}$$

and

$$\begin{cases} \cos\Delta_1 = 1-\dfrac{1}{\cos^2(2\varphi_1)}\left(1-\dfrac{1}{2}\sqrt{2(1+T_1)}\right) \\ \cos\Delta_2 = 1-\dfrac{1}{\cos^2(2\varphi_2)}\left(1+\dfrac{1}{2}\sqrt{2(1+T_1)}\right) \end{cases} \qquad \text{(11a and b)}$$

By means of equations (10) and (11) the fibril angle φ and the relative retardation Δ of fiber 3 can be calculated when $T_1 \neq \pm 1$. The calculation will yield two solutions for φ and Δ. Only one of them has physical significance, the other one is a useless

mathematical byproduct. One way to distinguish the physically relevant solution from the mathematical byproduct is to repeat the same measurement with another wavelength. It is based on the fact that for a linear retarder the orientation angle of the optical axis or the fibril angle φ in this description is independent of wavelength, while the relative retardation Δ is related to the used wavelength λ according to the relationship (see e.g. Amnon Yariv and Pochi Yeh)

$$\Delta = \frac{360°d(n_1-n_2)}{\lambda}, \qquad (12)$$

where d is the fiber-wall thickness, n_1 and n_2 the refractive indices for light vibrations parallel and perpendicular to the microfibril direction, respectively, and n_1-n_2 is the birefringence of the wall material.

Equations (10) and (11) are not valid in the case of $T_1=\pm1$. The special cases of $T_1=\pm1$ should be avoided, because the amplitude of the sinusoidal dependence I(A) reaches its maximum when $T_1 =\pm1$ ($T_2=0$ in this case). For the PSA (Polarizer-sample-analyzer)-arrangement of the intensity-quotient method (ref. Holzapfel and Ye) errors rapidly increase when the amplitude of I(A) will be equal or nearly equal to one. The method of the invention is based on the intensity-quotient principle and its measurement arrangement is also a PSA system. Therefore, the error of the method of the invention will increase rapidly, in a way similar to that of the PSA arrangement of the intensity-quotient method, when the amplitude of I(A) approaches one. A such critical case can be avoided by changing the wavelength used.

Using the method of the invention, single pulp fibers have been measured to demonstrate it. Because all the existing polarized-light methods are applicable only to one-wall measurement, they cannot be easily used for testing the two-wall method of the invention. To test the new method the multi-wavelength measurement principle was employed based on equa-

tion (12) and the fact that the fibril angle is wavelength-independent. For example, in the case of two wavelengths λ_1 and λ_2, two groups of intermediate results, say φ_{11}, Δ_{11} and φ_{12}, Δ_{12} for λ_1 and φ_{21}, Δ_{21} and φ_{22}, Δ_{22} for λ_2 are obtained. If φ_{11}, Δ_{11} and φ_{21}, Δ_{21} are the physically meaningful results, the relations $\varphi_{11} \approx \varphi_{21}$ and $\Delta_{11}/\Delta_{21} \approx \lambda_2/\lambda_1$ should be valid according to the just mentioned criteria. On the contrary, being mathematical byproducts the rest cannot meet them. On the other hand, the method should be effectively demonstrated, if the two criteria described above are satisfactorily fulfilled, especially in a broad wavelength bandwidth.

The experimental setup has the same configuration as shown in Fig. 2. In the test experiments the wavelength λ of the incident light beam was changed from 400-700 nm with a step of 50 nm. Single pine kraft pulp fibers were employed as the test samples. For each test fiber a straight and nondamaged segment was selected and the intensity I(A) of two points in its centre region was detected. The intensity I(A) was measured by rotating the analyzer from A=-45° to A=-225° every 15° for each wavelength. From the intensity data I(A=-45°), I(A=-90°), I(A=-135°) and I(A=-180°), which are equivalent to I(A=135°), I(A=90°), I(A=45°) and I(A=0°), φ and Δ were calculated using equations (9), (10) and (11). For each wavelength two groups of values for φ and Δ were obtained. From the calculated intermediate results, as theoretically predicated, the physically meaningful ones could be easily distinguished by using the two criteria given above. The present invention will be explained in more detail with reference to the following examples which are the results of some demonstration measurements.

EXAMPLE 1

The first example was unbleached commercial pine kraft pulp (FIG. 3a). FIG. 3(b) is the magnified picture of the measured fiber. Figures 4a), 4b), 4c) and 4d) show the measured results of φ and Δ of the fiber at point 1 and point 2 (marked

in FIG. 3b) versus wavelength λ, respectively. As expected, the φ values obtained do not differ very much from each other (see FIG. 4a and FIG. 4c). The average of all measured φ values is equal to -17.67° for point 1 and -18.90° for point 2. The minus sign in the coordinate system of the experiments means that the helix formed by the microfibrils of the S_2 layer is right-handed. In FIGS. 4b and 4d the measured data for Δ are denoted by rectangles with the dashed curve drawn only to illustrate the data. Also as expected, the Δ values are increased with reduced wavelength λ. The regression curves indicated by circles with a solid curve were fitted based on the measured data and by using equations (7), (8) and (12). It can be seen that the measured Δ values coincide well with the fitting curve.

With all measured data of I(A) the least square method was employed to determine φ and Δ. For example, FIG. 5 shows the measured relative intensity change $I(A)/I_0$ from A=-45° to A=-225° at point 1 of the fiber in FIG. 3b under the illumination of λ=600 nm. The differences between the measured data and their sine regression curve fitted by using equations (7) and (8) are very small and the sum of the squares of the differences amounts 0.0017. The best estimates for φ and Δ based on all the data of I(A) and determined by using the least square fitting are not significantly different from those obtained by using the four intensity values.

EXAMPLE 2

The second sample was unbleached laboratory-made pine kraft pulp (FIG. 6a), and one pulp fiber (FIG. 6b) was selected for the test measurement. The results obtained for φ and Δ of the fiber at point 1 and point 2 (marked in FIG. 6b) in the wavelength range 400-550 nm are given in FIGS. 7a, 7b, 7c and 7d, respectively. The results for the range 600-700 nm due to relatively large errors were not accepted. To find out the cause for the large errors, the intensity data was examined closer and it was found that the sine intensity curves I(A) obtained when λ=600-700 nm really have a very large amplitude.

As pointed out, the error of the method of the invention increases rapidly, in a way similar to that of the PSA arrangement of the intensityquotient method, when the amplitude of I(A) approaches one. For a pulp fiber the fibril angle φ remains constant and the retardation Δ changes when the wavelength λ varies (ref. equation (12)). At certain wavelengths retardation values will become so large that I(A) has a large amplitude and the error increases rapidly. The results for the range λ=600-700 nm obviously reflect this case. Practically the measurement of this fiber in the range of 400-550 nm has indicated that it is always possible to avoid a such critical case by changing the wavelength used.

EXAMPLE 3

FIG. 8a shows the third sample, a bleached commercial pine kraft pulp, and FIG. 8b the measured fiber segment. The φ and Δ obtained for point 1 and point 2 (marked in FIG. 8b) are shown in FIGS. 9a, 9b, 9c and 9d as a function of the wavelength in the range 400-700 nm. The results of point 1 for this fiber show almost perfect agreement with the theoretical expectations both for φ and Δ (FIGS. 9a and 9b).

What we claim is:

1.A method for determining the orientation angle of the optical axis and the relative phase retardation of a birefringent specimen that can be described by two linear retarders having the same retardation in cascade, whose optical axes are crossed, the method comprises the steps of:

producing a linearly polarized light beam at at least two sequential predetermined wavelengths;

impinging the linearly polarized light beam on the specimen to be measured;

orienting the axis of the specimen or the bisection of its two optical axis directions at a predetermined angle different from zero or 90° relative to the polarization plane of the incident linearly polarized light beam;

measuring the light intensity emergent from the specimen by rotating an analyzer located behind the specimen for each wavelength, respectively;

calculating the intermediate results for the orientation angle of the optical axis and the relative phase retardation of the specimen under test for each wavelength; and

determining the orientation angle of the optical axis and the relative phase retardation of the specimen from the all intermediate results obtained by comparing them with each other and taking into account that the orientation angle of the optical axis is independent of wavelength, whereas the relative phase retardation is inversely proportional to the wavelength used.

2. A method for determining the fibril angle and the relative phase retardation of single, intact pulp fibers comprising the steps of:

producing a linearly polarized light beam at at least two sequential predetermined wavelengths;

impinging the linearly polarized light beam on the pulp fiber to be measured;

orienting the axis of the fiber relative to the polarization plane of the incident linearly polarized light beam;

measuring the light intensity emergent from the fiber under test by rotating an analyzer located behind the fiber;

calculating and determining the fibril angle and the relative phase retardation of the fiber under test based on the intensity data obtained.

3. The method as claimed in claim 2, wherein the linearly polarized light of one wavelength is impinged on the fiber to be measured and the fiber to be measured is oriented with its axis at a predetermined angle different from zero or 90° related to the polarization plane of the incident linearly polarized light.

4. The method as claimed in claim 2, wherein the light intensity I(A) emergent from the fiber to be measured behind said analyzer is detected at the four analyzer positions of azimuth A=0°, 45°, 90° and 135° or their corresponding periodic positions with the fiber axis as the coordinate axis, and wherein the intensity quotients $(I(45°)-I(135°))/(I(45°)+I(135°))$ and $(I(0°)-I(90°))/(I(0°)+I(90°))$ are calculated and with the intensity quotients one group of intermediate results for the fibril angle and the relative phase retardation of the fiber is obtained.

5. The method as claimed in claim 2, wherein the wavelength of the incident linearly polarized light is changed, the measurement and data-processing processes are repeated for this

wavelength and another group of intermediate results for the fibril angle and the relative phase retardation is obtained.

6. The method as claimed in claim 2, wherein from the all intermediate results for the fibril angle and the phase retardation obtained with two wavelengths the final measurement results are determined by comparing them with each other and taking into account that the fibril angle is independent of wavelength, whereas the relative phase retardation is inversely proportional to the wavelength used.

7. The method as claimed in claim 2, wherein the measurement and data-processing processes are repeated for more than two wavelengths, more than two groups of intermediate results for the fibril angle and the phase retardation are obtained, and from the all intermediate results the final measurement results for the fibril angle and the phase retardation are determined according to the judgement principle that the fibril angle is independent of wavelength, whereas the relative phase retardation is inversely proportional to the wavelength used.

8. The method as claimed in claim 2, wherein the light intensity I(A) emergent from the fiber to be measured behind said analyzer is detected by rotating said analyzer or changing the azimuth A of said analyzer relative to the axis of said fiber in such a way that the intensity I(A) within at least one whole period is registered, and wherein the intermediate results for the fibril angle and the phase retardation are obtained by generating sine curve for I(A) that fits the measured intensity data of I(A) by varying the estimation values for the fibril angle and the phase retardation based on the least square principle.

9. The method as claimed in claim 8, wherein the measurement and data processing processes are repeated for two or more than two wavelengths, and wherein two or more than two groups of intermediate results for the fibril angle and the phase

retardation are obtained and from the all intermediate results the final measurement results for the fibril angle and the phase retardation are determined according to the judgement principle that the fibril angle is independent of wavelength, whereas the relative phase retardation is inversely proportional to the wavelength used.

10. A device for determining the fibril angle and the relative phase retardation of single, intact pulp fibers comprising

a light source means for sequentially generating a light beam of at least two wavelengths;

a polarization-optical image system for generating the polarizing micrographs of the fiber under test required for calculating its fibril angle and relative phase retardation;

a detector means for recording the light intensity emergent from said polarization-optical image system; and

means for image-processing and data-processing, and wherein said light source means, polarization-optical image system and detector are oriented in cascade to build up an image polarimeter or ellipsometer.

11. The device as claimed in claim 10, wherein said light source means can be either an optical monochromator or a tunable laser that provides monochromatic or quasi-monochromatic, preferably visible, radiation of at least two wavelengths or a normal polychromatic light source equipped with a set of at least two wavelength filters so that radiation of at least two wavelengths can be produced by placing one of the filters into the optical path and exchanging it with the others or a monochromatic or quasi-monochromatic light source equipped with a tunable filter, e.g. acousto-optical tunable filter, so that the wavelength of its output radiation can be changed.

12. The device as claimed in claim 10, wherein said polarization-optical image system comprises a polarizer that linearly polarizes the monochromatic or quasi-monochromatic light emergent from said light source means, a workstage on which the pulp fiber to be measured is put, a condenser lens for focusing the linearly polarized light existing on the fiber under test, an objective lens installed behind the workstage to produce a magnified fiber image and a rotatable analyzer, and wherein the axis of the fiber to be measured is oriented at a certain angle different from zero or 90° relative to the transmission axis of said polarizer.

13. The device as claimed in claim 12, wherein said work-stage is replaced by a sampling system with a capillary lying in a plane perpendicular to the light propagation direction between said polarizer and analyzer, and wherein said sampling system is oriented so that the axis of said capillary makes a certain angle, most preferably an angle of 45°, to the transmission axis of said polarizer and the fibers to be measured are one by one passed through said capillary.

14. The device as claimed in claim 12, wherein said polarizer and analyzer are substituted by their corresponding fiber-optical and/or integrated optical elements.

15. The device as claimed in claim 10, wherein said detector means is a CCD camera or microscope video camera in conjunction with a camera control unit and said CCD camera or microscope video camera is interfaced to said image-processing unit where the intensity data detected by said CCD camera or microscope video camera are digitized and processed.

16. The device as claimed in claim 10, wherein said means for image-processing and data-processing consists of a frame grabber, a computer and an image analysis program.

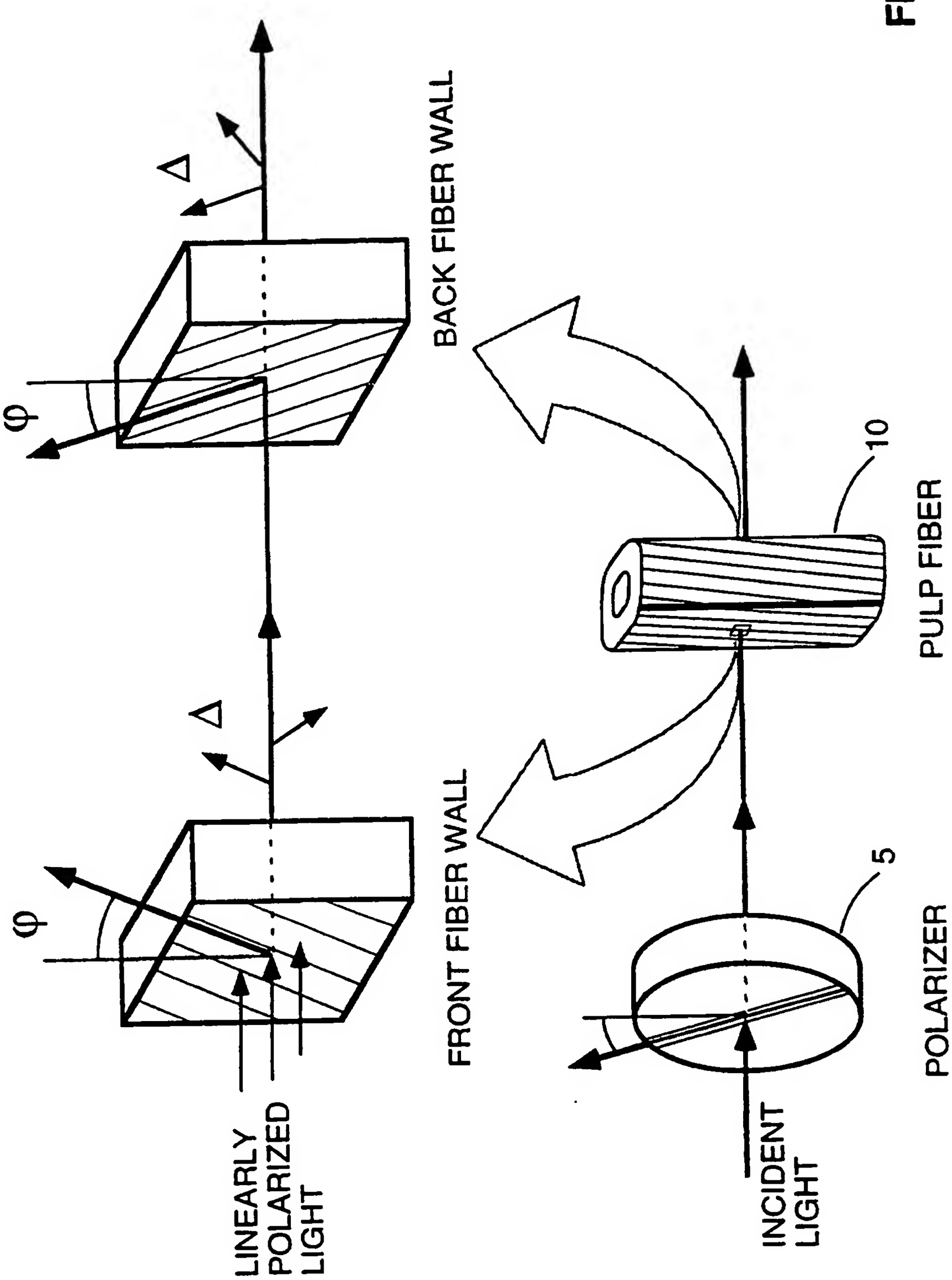
LINEARLY POLARIZED LIGHT
φ
Δ
FRONT FIBER WALL
BACK FIBER WALL
PULP FIBER
10
POLARIZER
5
INCIDENT LIGHT

FIG. 1

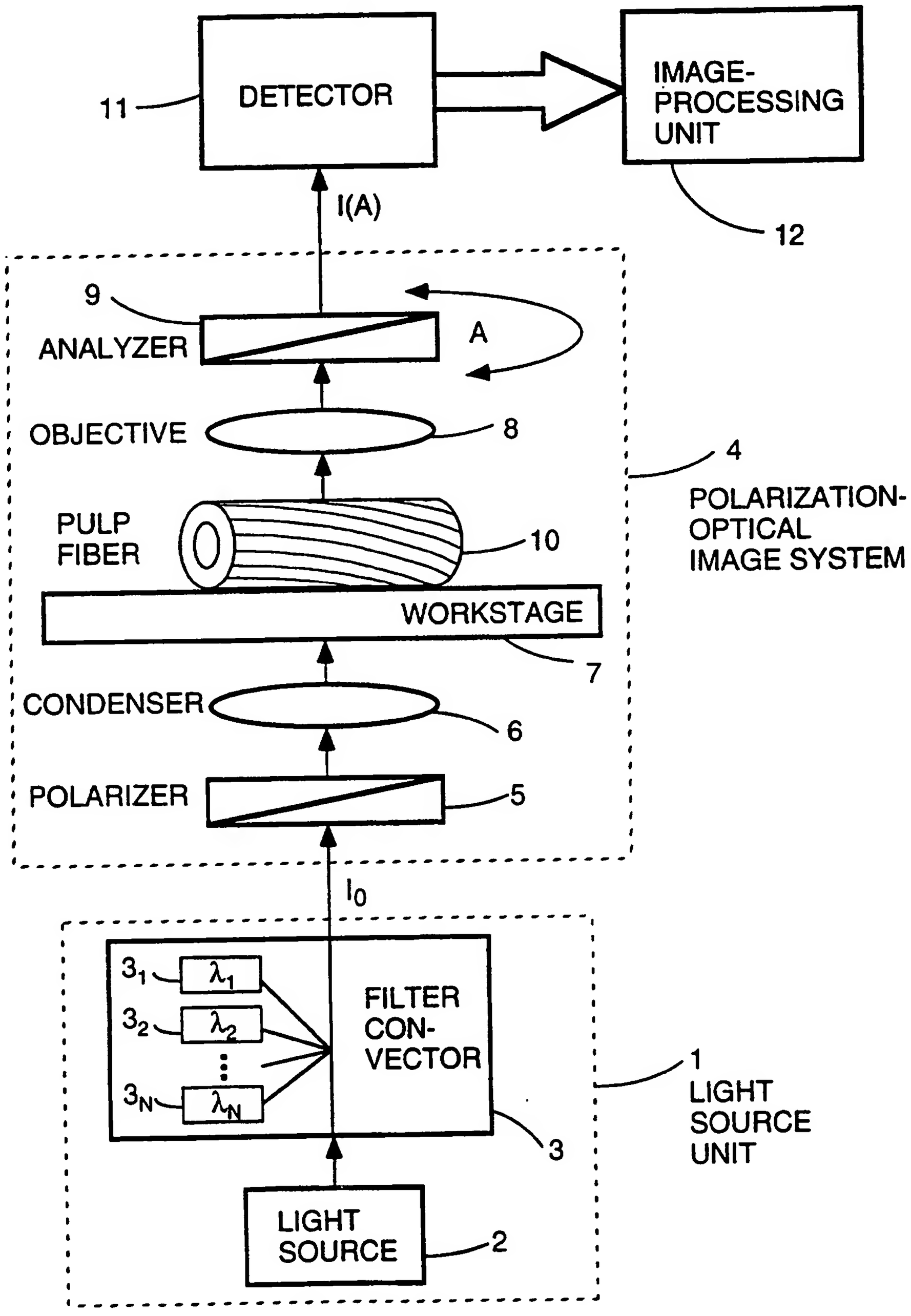
11
DETECTOR
IMAGE-
PROCESSING
UNIT
12
I(A)
9
ANALYZER
A
OBJECTIVE
8
4
POLARIZATION-
OPTICAL
IMAGE SYSTEM
PULP
FIBER
10
WORKSTAGE
7
CONDENSER
6
POLARIZER
5
I_0
3_1
λ_1
3_2
λ_2
3_N
λ_N
FILTER
CON-
VECTOR
3
1
LIGHT
SOURCE
UNIT
LIGHT
SOURCE
2

FIG. 2

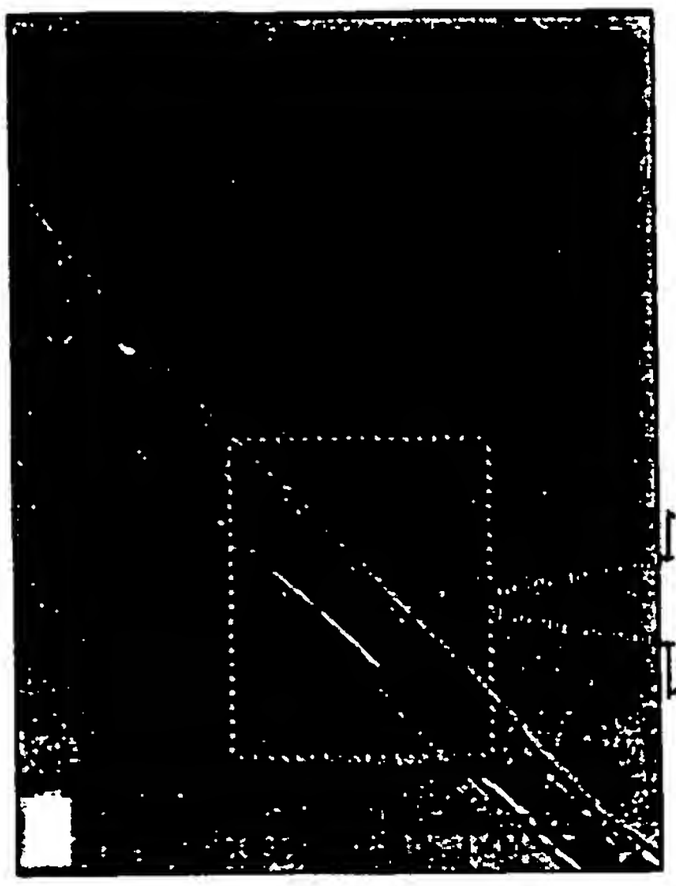

FIG. 8A

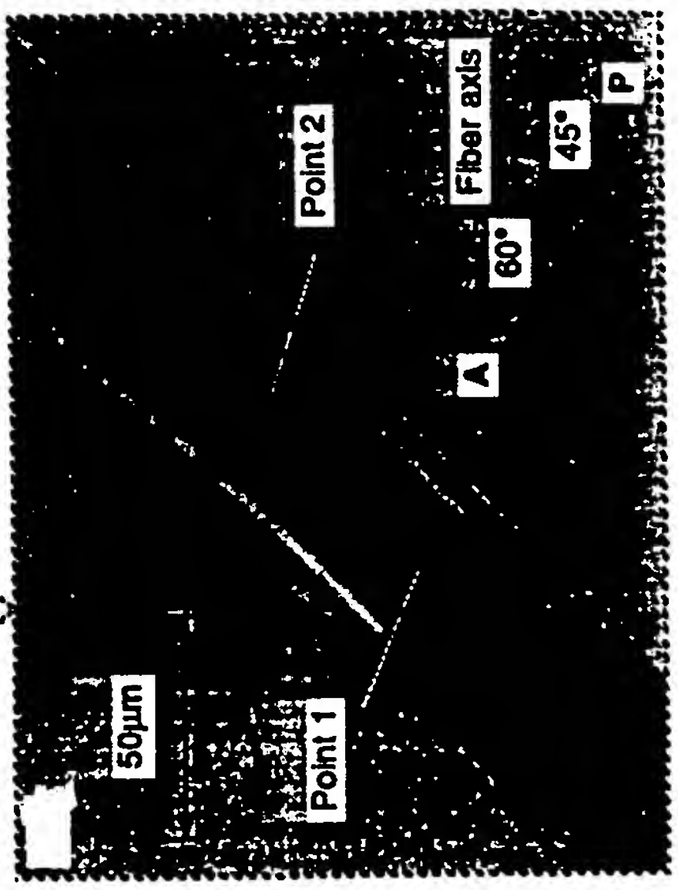

FIG. 8B

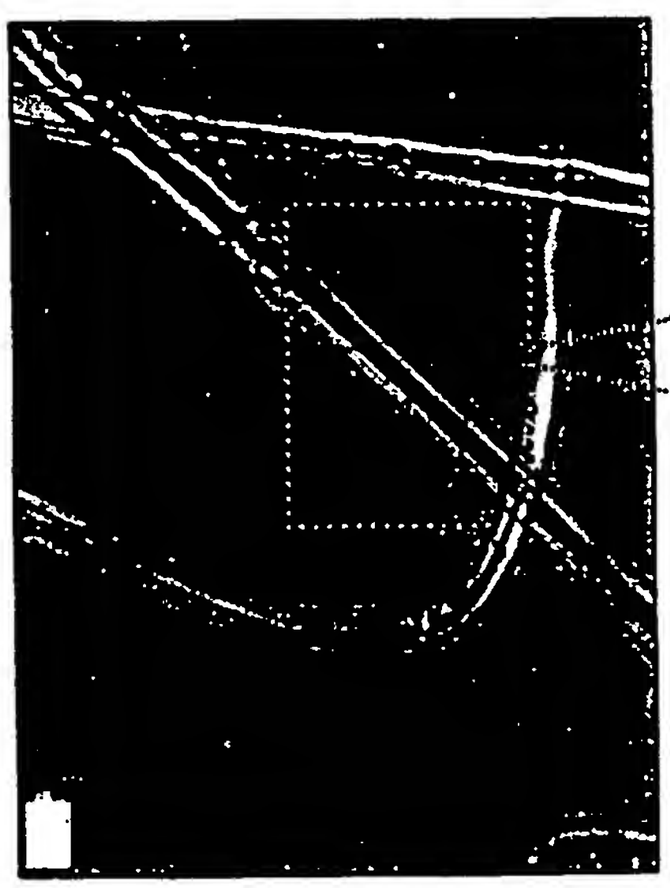

FIG. 6A

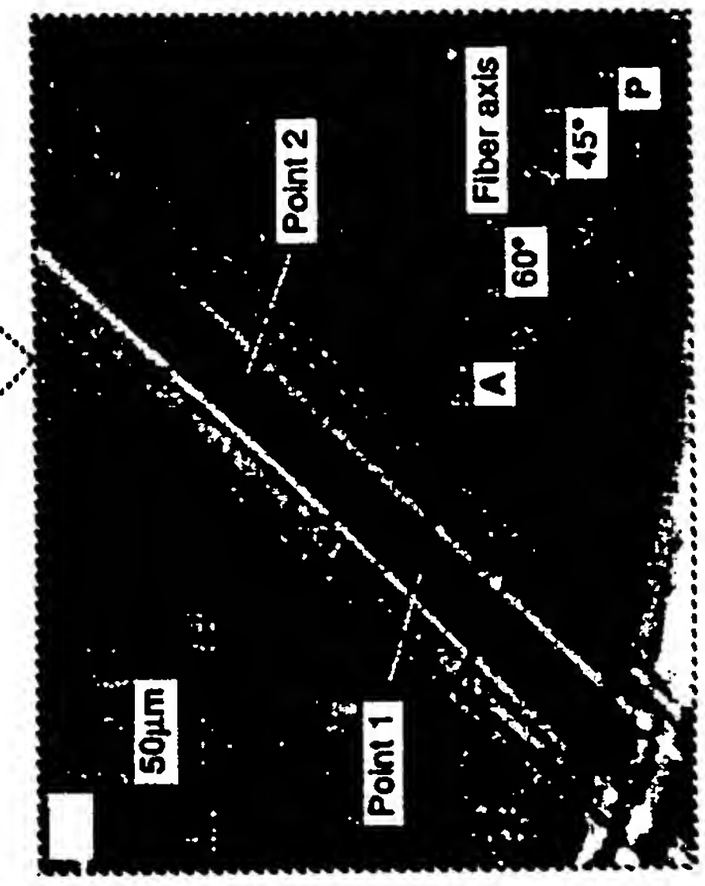

FIG. 6B

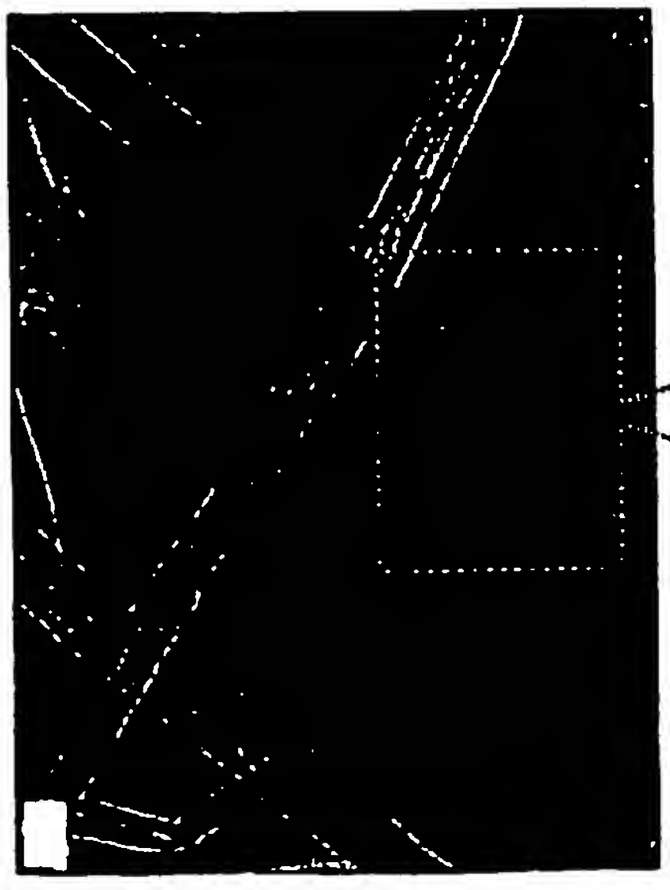

FIG. 3A

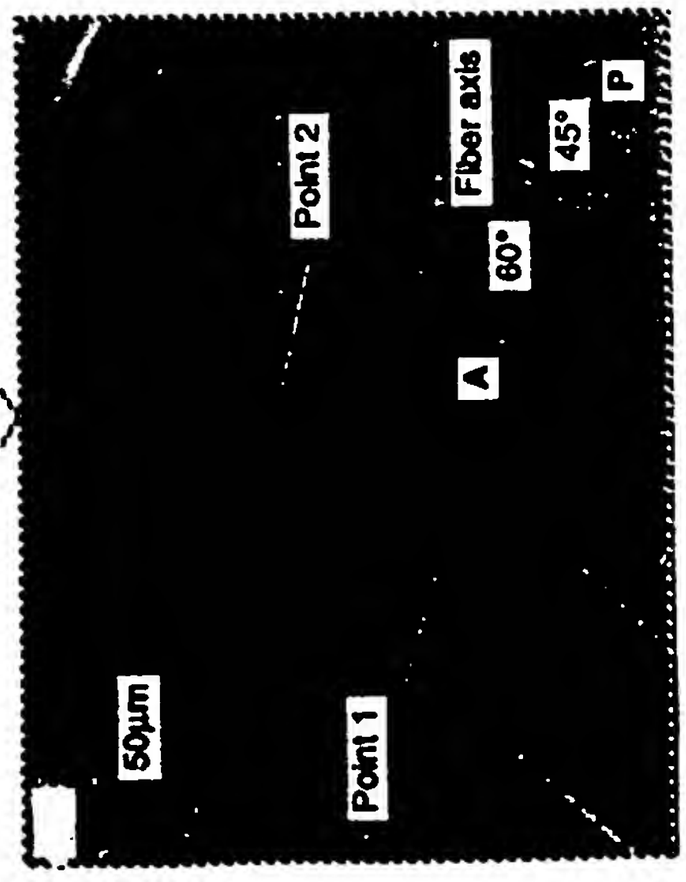

FIG. 3B

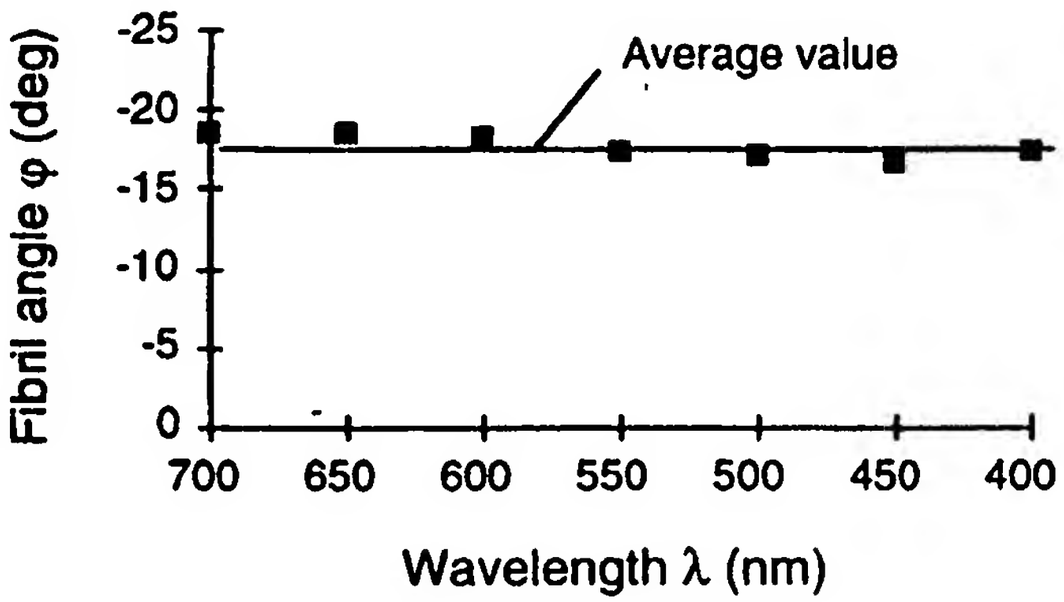

FIG. 4A

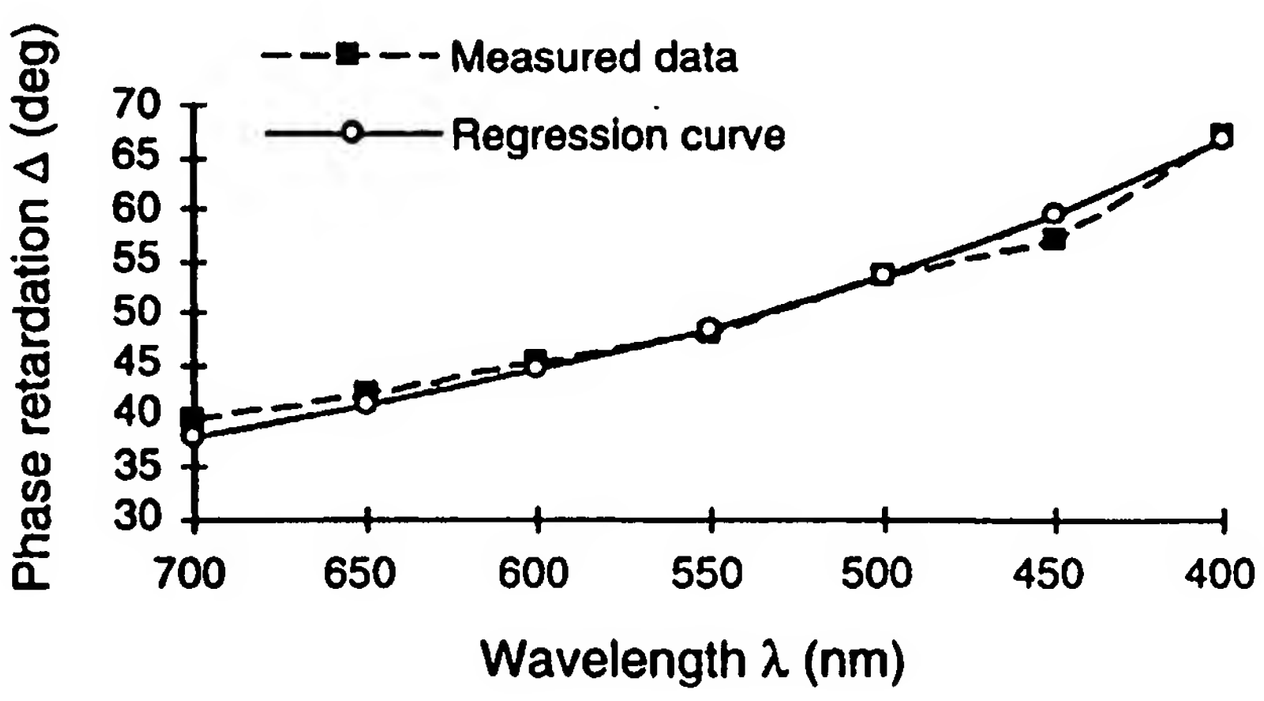

FIG. 4B

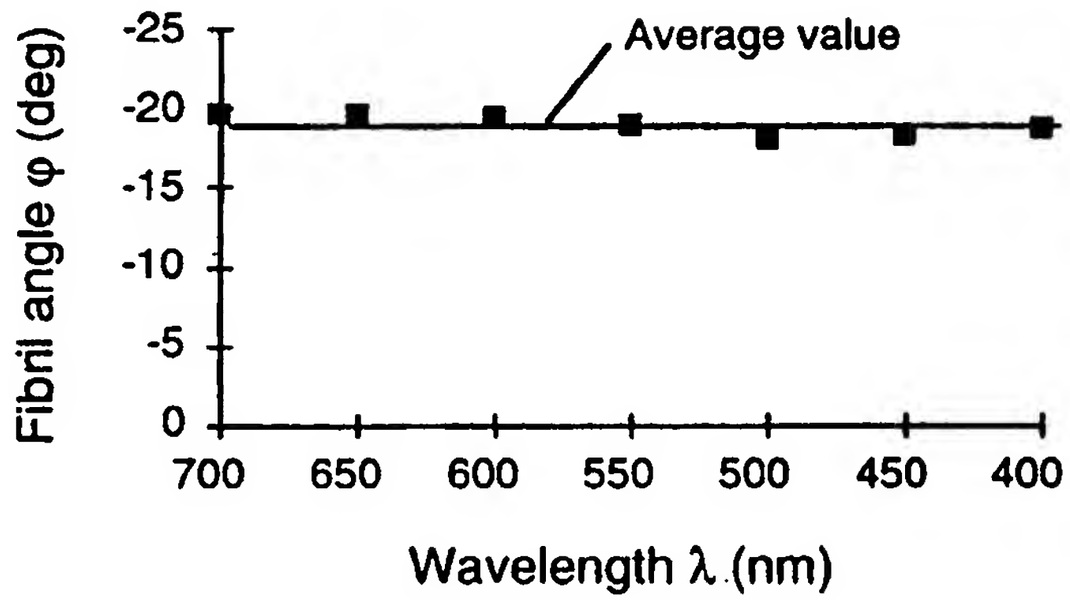

FIG. 4C

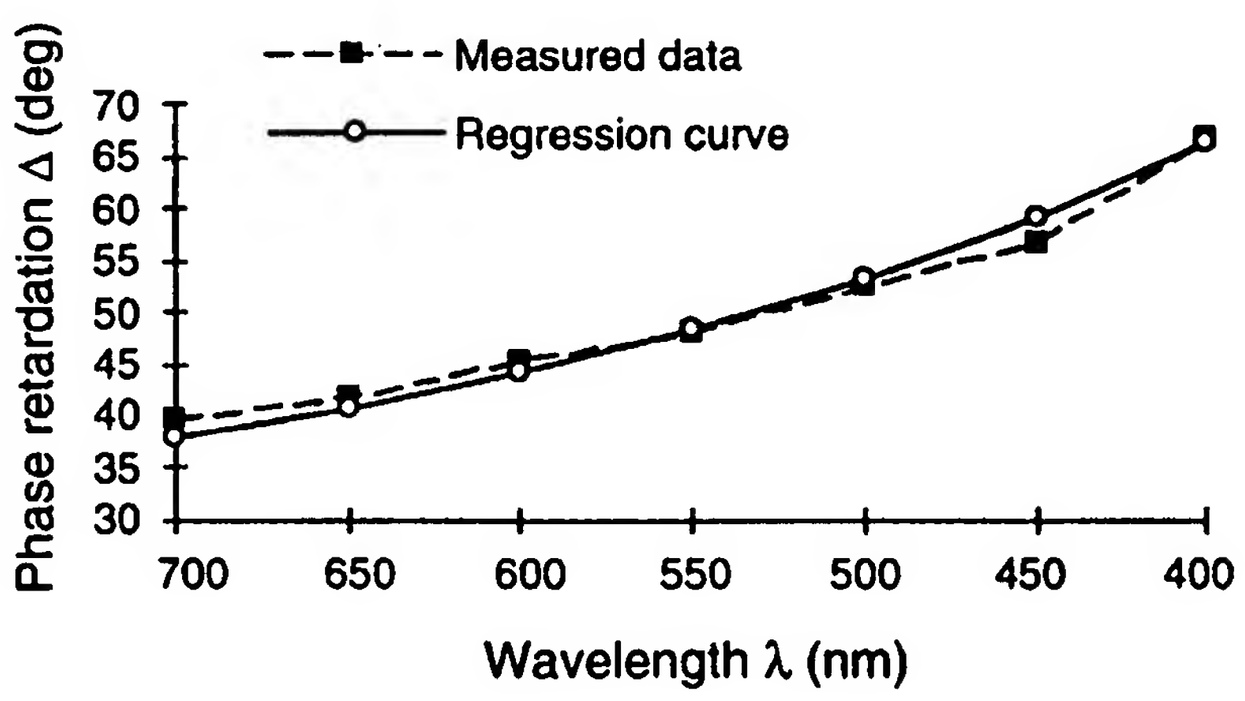

FIG. 4D

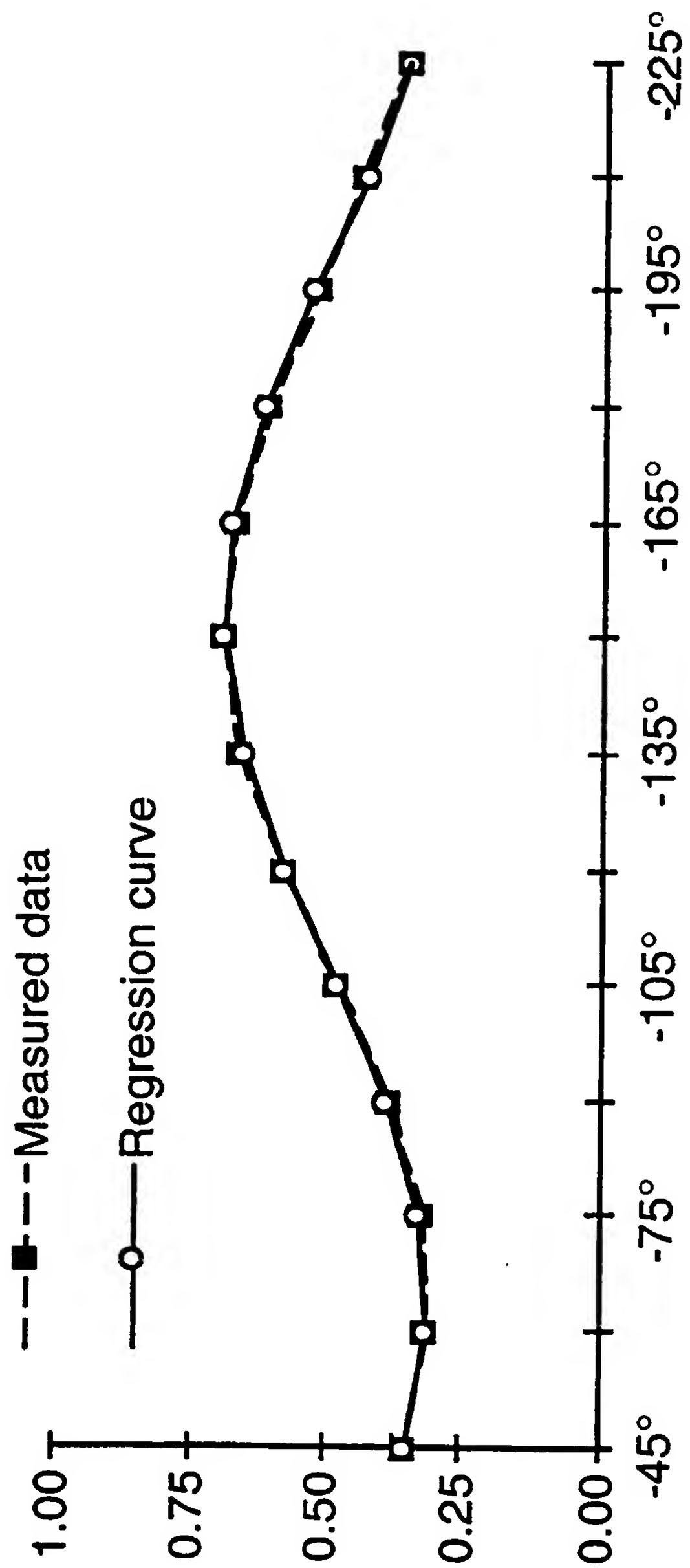
Relative intensity I(A)/I0
1.00
0.75
0.50
0.25
0.00
-45°
-75°
-105°
-135°
-165°
-195°
-225°
Analyzer azimuth A
Measured data
Regression curve

FIG. 5

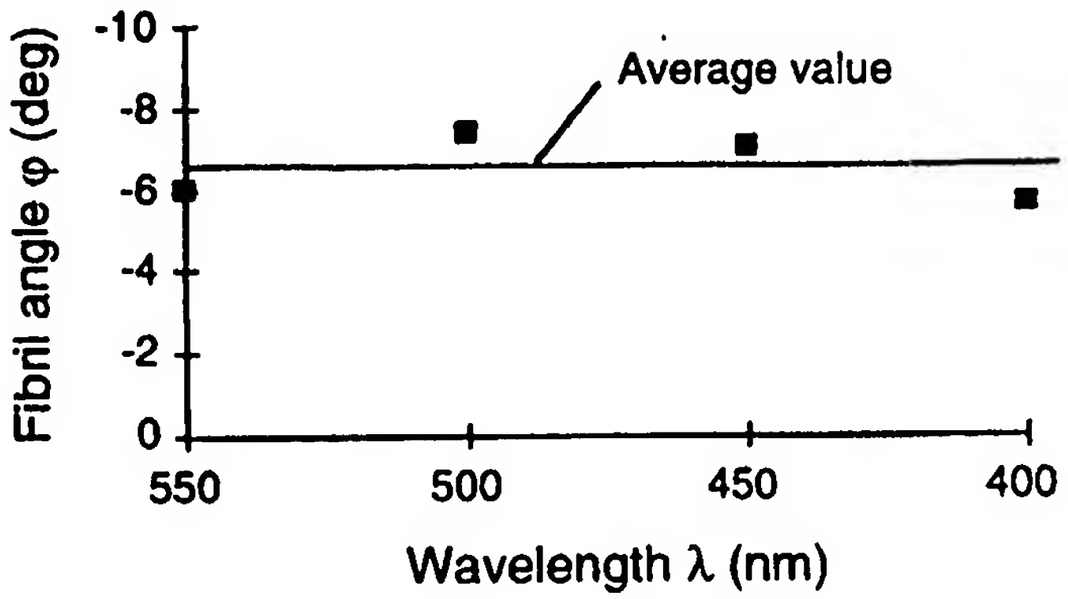

FIG. 7A

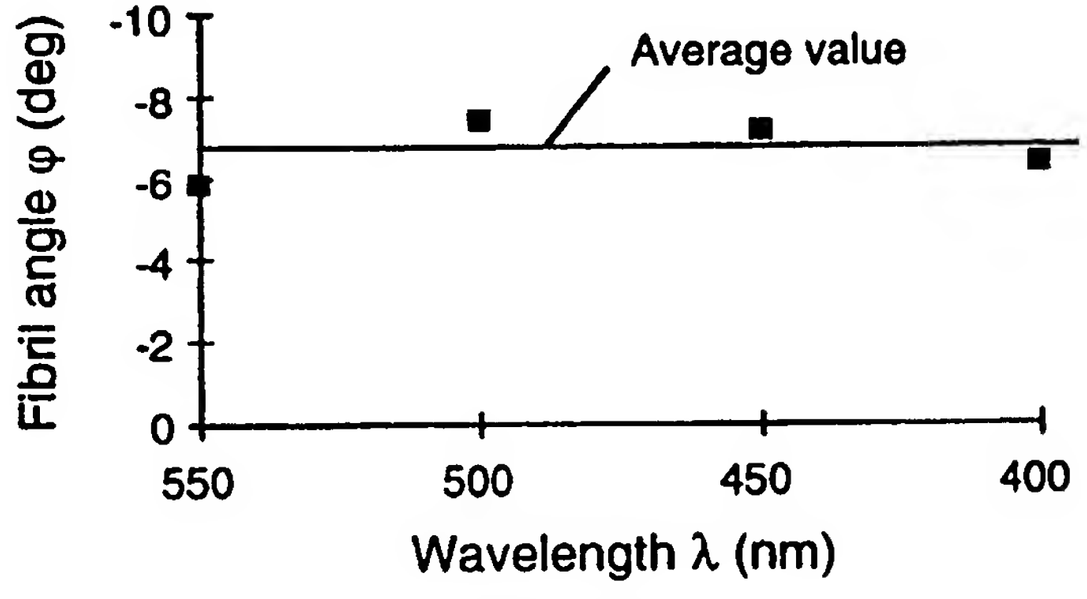

FIG. 7C

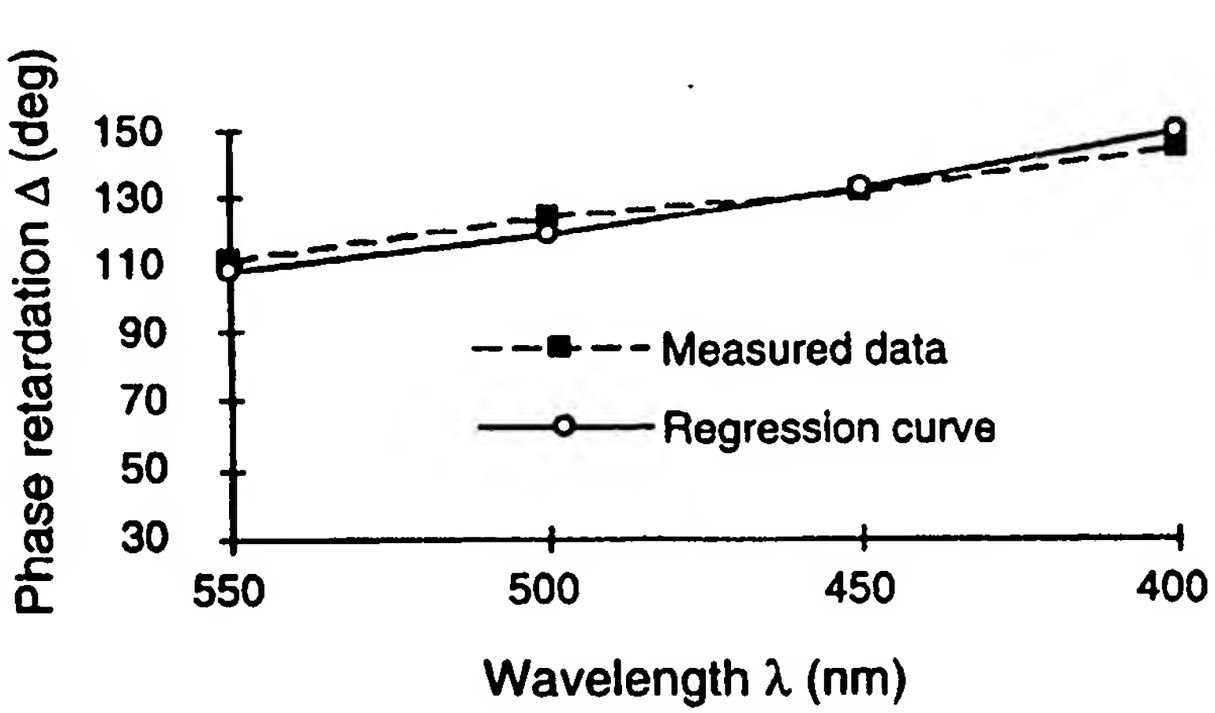

FIG. 7B

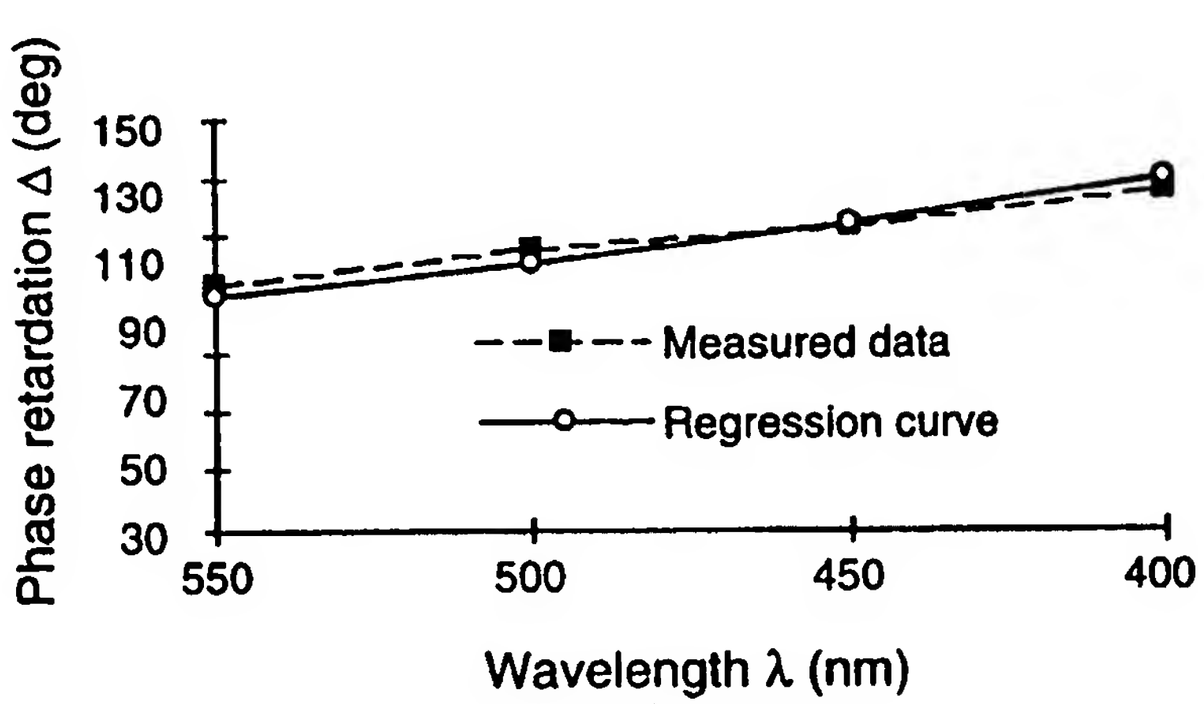

FIG. 7D

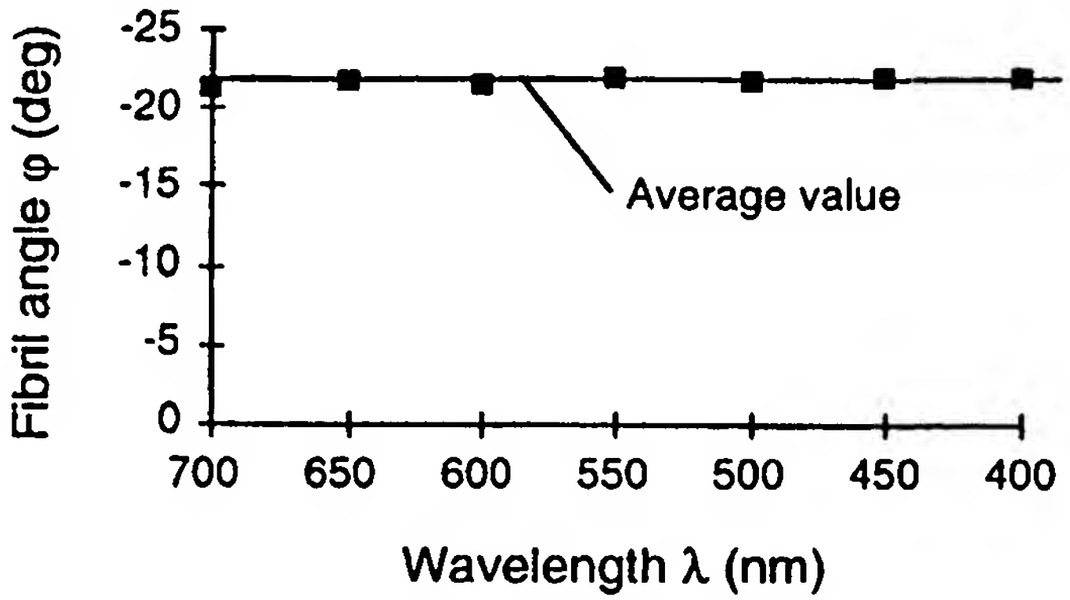

FIG. 9A

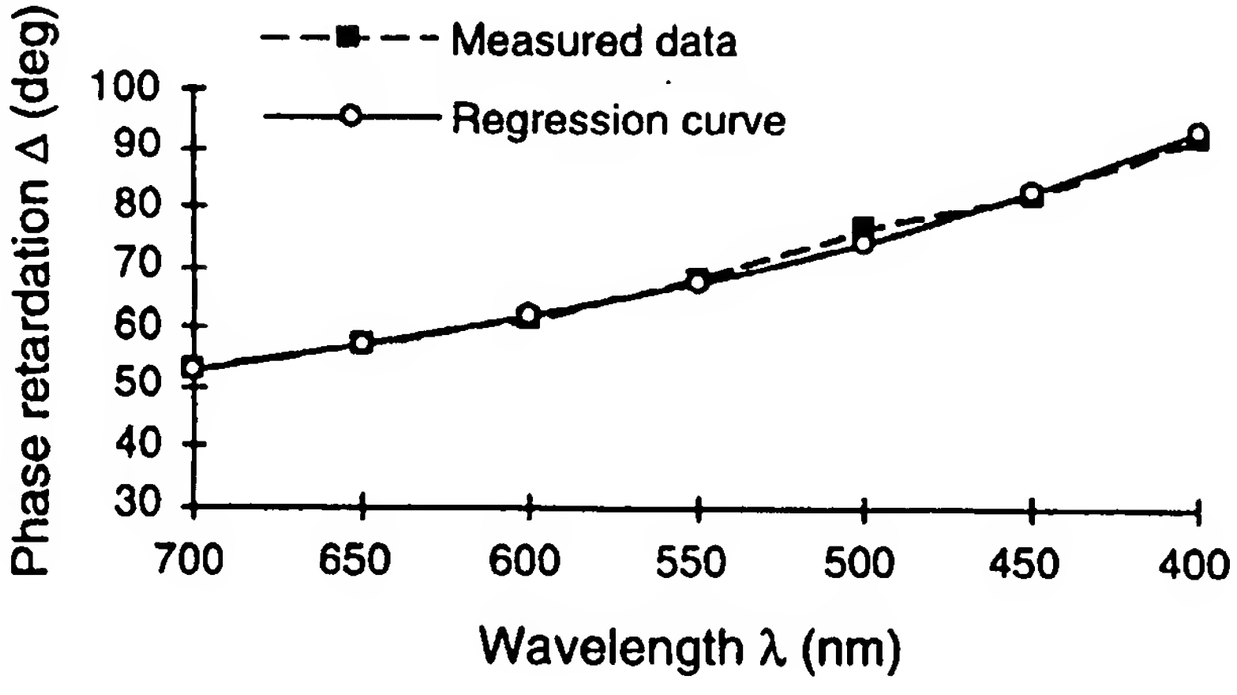

FIG. 9B

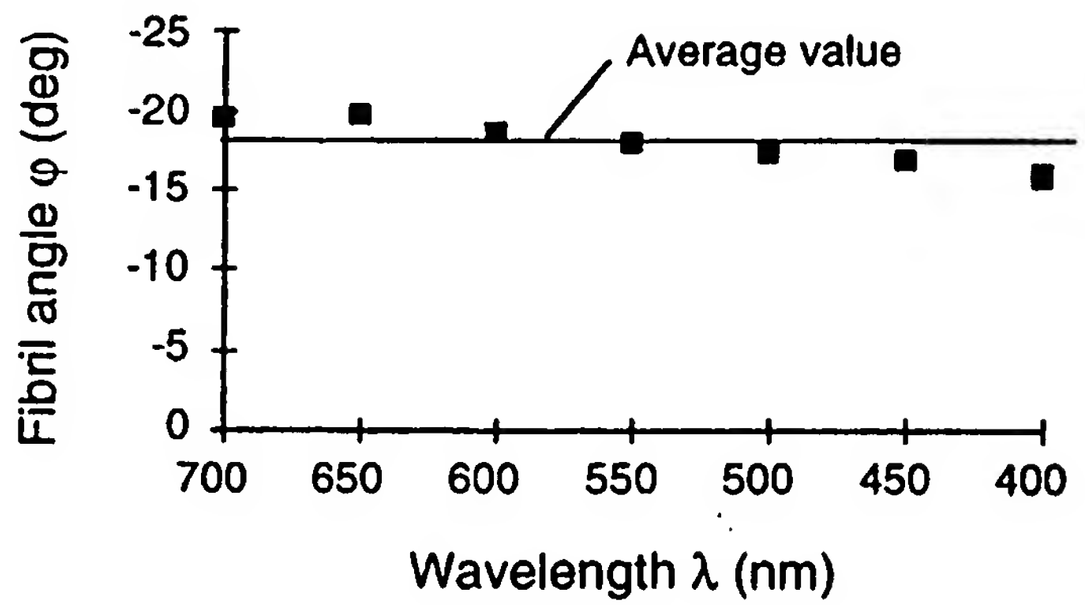

FIG. 9C

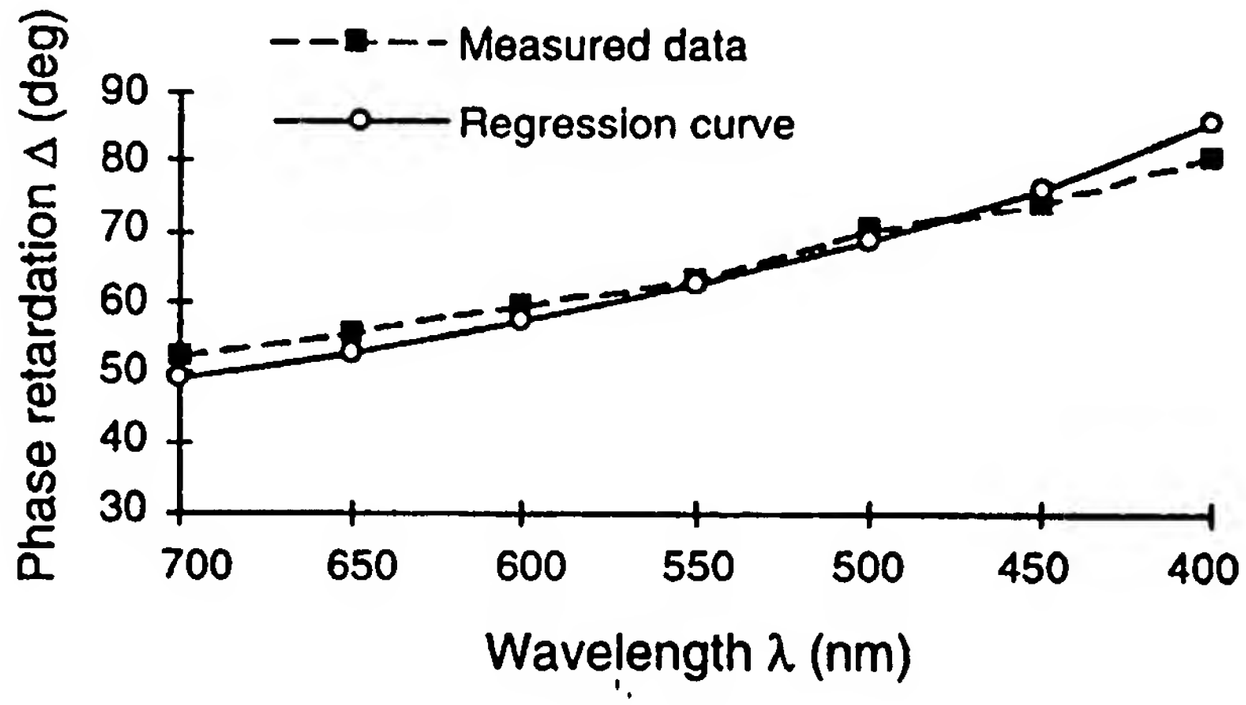

FIG. 9D

INTERNATIONAL SEARCH REPORT

International application No.
PCT/FI 95/00531

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: G01N 21/21, G01J 3/447
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: G01N, G01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, CLAIMS

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4973163 A (K. SAKAI ET AL), 27 November 1990 (27.11.90), column 3, line 64 - column 4, line 19, figure 1, abstract ---	1-16
A	EP 0160304 A2 (KANZAKIPAPER MANUFACTURING COMPANY LIMITED), 6 November 1985 (06.11.85), page 6, line 33 - page 8, line 20, figure 1, abstract ---	1-16
A	US 5087823 A (J. SILVY ET AL), 11 February 1992 (11.02.92), column 3, line 63 - column 4, line 17; column 5, line 32 - column 6, line 55, abstract ---	1-16

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" erlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
6 February 1996	08.02.96
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer Karin Säfsten Telephone No. +46 8 782 25 00

INTERNATIONAL SEARCH REPORT

International application No.
PCT/FI 95/00531

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4171916 A (R.J. SIMMS ET AL), 23 October 1979 (23.10.79), column 2, line 29 - column 3, line 56, figure 1, abstract -- --------	1-16

INTERNATIONAL SEARCH REPORT

Information on patent family members

05/01/96

International application No.
PCT/FI 95/00531

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A- 4973163	27/11/90	JP-A- 2102436	16/04/90
		JP-A- 2102437	16/04/90
EP-A2- 0160304	06/11/85	JP-A- 60231136	16/11/85
US-A- 5087823	11/02/92	CA-A- 2014388	12/10/90
		DE-D- 69005009	00/00/00
		EP-A,B- 0392943	17/10/90
		FR-A,B- 2645961	19/10/90
US-A- 4171916	23/10/79	CA-A- 1116435	19/01/82
		JP-A- 54078189	22/06/79
		SE-A- 7811882	19/05/79